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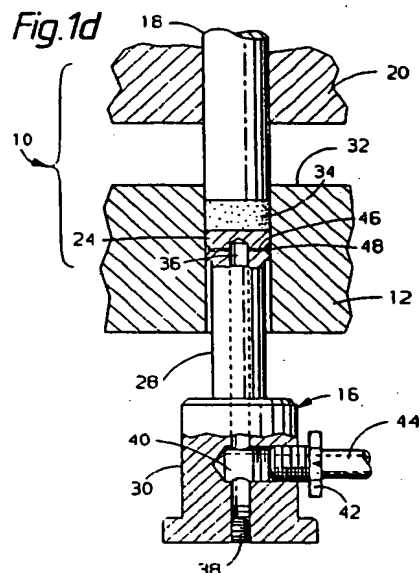
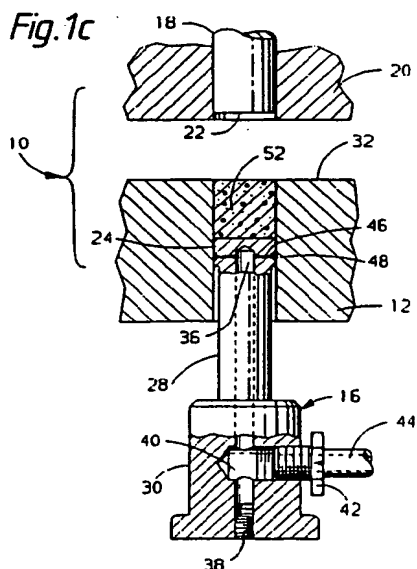
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(54) Method of producing magnetic bodies using a lubricated cold-press die.

(57) A rare earth-containing powder (52) is pressed into a compact body (34) at ambient temperatures using a solid lubricant applied only to the die wall

and/or a core. The solid lubricant is applied by suspending lubricant powder in a fluorinated hydrocarbon liquid.

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This invention pertains to practices for the hot-pressing of rare earth element-containing powder alloys. More particularly, this invention pertains to a lubrication practice for forming a cold-pressed compact body as specified in the preamble of claim 1.

Rare earth element-containing alloys composed so as to form a  $\text{RE}_2\text{TM}_{14}\text{B}$  tetragonal crystal phase have been melt-spun under carefully controlled processing to produce useful permanent magnet materials as disclosed in US-A-4,802,931 and US-A-4,851,058. Such melt-spun materials either as quenched or in an overquenched and annealed condition consist essentially and predominantly of a tetragonal crystal, prototype  $\text{Nd}_2\text{Fe}_{14}\text{B}$  phase. The tetragonal crystal-containing grains are very small, typically less than a few hundred nanometres on the average in grain size, and are surrounded by one or more secondary grain boundary phases which contribute to the permanent magnet characteristics of the composition. This fine grain material is magnetically isotropic, and the melt-spun ribbon fragments can be pulverized to a suitable powder, combined with a suitable binder material and moulded into useful bonded isotropic permanent magnets as disclosed in US-A-4,902,361.

Where permanent magnets of higher energy product are desired, it is known that the melt-spun powder material can be hot-pressed to form a fully-densified permanent magnet body and that, where desired, such a fully-densified body can be further hot-work-deformed into a magnetically very strong, anisotropic magnet. These practices are disclosed, for example, in US-A-4,792,367 and US-A-4,844,754.

The fine-grain, melt-spun, rare earth element-containing material is initially in the form of ribbon particles or a powder produced by comminution of the ribbon fragments. In order to hot-press or otherwise hot-work the material, it is necessary that it should be heated to a suitable hot working temperature typically in the range of  $700^\circ\text{C}$  to  $800^\circ\text{C}$ . As disclosed in the aforesaid U.S. patents, it is prudent to heat the powder in vacuum or a suitable inert gas that provides a dry and substantially oxygen-free environment in order to prevent the powder from burning. In attempting to work with such readily oxidizable rare earth element-containing materials, it has been necessary to provide a suitable protective atmosphere in which the rare earth and other constituents are not oxidized and the permanent magnetic properties of the materials are not degraded.

In our co-pending European application number is disclosed a two-step cold-pressing-hot-pressing process for producing hot-pressed rare earth-transition metal-boron (RE-TM-B) magnets in

an open-to-the-air press. In the first step of that process, fine-grain RE-TM-B material in powder form is compacted at ambient temperature in open-to-the-air presses. The cold-pressed compact body that is formed has a density of about 5 to 5.5 grams per  $\text{cm}^3$ , which is about 70 percent of the density of a fully-densified body of the same composition. In accordance with such two-step practice, the cold compact bodies are then suitably hot-pressed in an open-to-the-air hot press in which the die cavity is heated and flooded with a dry inert gas such as argon to protect the compact body from burning or from other oxidation which would degrade the magnetic properties of the product.

The rare earth element-containing powder and cold-pressed compact body are both susceptible to reaction with moisture and with certain chemical species such as the chloride ion. Therefore, in order to prevent chemical reaction of the constituents of the powder or the compact body, it has been necessary to take precautions in addition to the use of dry inert gas during the hot-pressing operation. For example, in the cold-press-hot-press practice referred to above, a lubricant is used in the cold-pressing operation to facilitate compaction of the powder and removal of the compact body from the die without abrasion of the die or the compact body and without causing the compact body to split apart. In order to accomplish this successful cold-pressing, it was determined that a solid lubricant film, such as a film of polytetrafluoroethane or fluorinated ethylene-propylene copolymers (Teflon<sup>™</sup>), should be applied to the die wall. No lubricant should be mixed with the powdered material added to the die for compaction. If the compact was to be ring-shaped or the like and to require a core piece as part of the press tooling, a film of solid lubricant could also be suitably applied to the surface of such a core piece.

Teflon<sup>™</sup> powder is a preferred solid lubricant film. The application of the powder to the die or core surface is accomplished using a suspension of Teflon<sup>™</sup> in a volatile vehicle, preferably a liquid of relatively high density, so as to better suspend the Teflon<sup>™</sup> powder particles. Volatile chlorine-fluorine containing aliphatic hydrocarbon liquids have been used in the above-described process to suspend the Teflon<sup>™</sup> particles. However, some liquid remains in the lubricant film after drying and is transferred to the cold compact body. In some operations, it is necessary or desirable to store cold compact bodies for hours or days before they are hot-pressed. During such time, trace amounts of chlorine-containing liquids or other reactive liquids, particularly humid atmospheres, can react with the rare earth element-containing powder. Such reaction degrades the permanent magnet properties of the resultant product such as by

reducing its magnetic coercivity.

Accordingly, it is an object of the present invention to provide a suitable liquid vehicle for suspending solid lubricant materials such as Teflon<sup>™</sup> powder for die lubrication in the making of rare earth element-containing cold compact bodies for subsequent hot pressing.

A method of cold-pressing a rare earth element-containing alloy powder according to the present invention is characterised by the features specified in the characterising portion of claim 1.

In accordance with preferred embodiments of the invention, the above and other objects are accomplished as follows.

The starting material for the practice of the invention is suitably a melt-spun ribbon particle or powder composition composed so as to ultimately form a magnet body consisting essentially of the tetragonal phase RE<sub>2</sub>TM<sub>14</sub>B and a minor portion of a grain boundary phase(s) of higher rare earth element content. Whilst RE stands for rare earth elements generally, it is preferred that the rare earth constituent of this material be made up of at least 60 percent of neodymium and/or praseodymium. The transition metal element (TM) is preferably iron or mixtures of iron with cobalt and/or with minor portions of other metals. This rapidly-solidified starting material will suitably be of very fine grain size (e.g., less than 50 nm) or almost amorphous. The hot-pressing process and any additional hot-working process will then densify and work the material and simultaneously effect a growth in grain size such that the average grain size is larger but still less than about 500 nm in largest dimension. The product has useful permanent magnet properties.

The practice of the present invention is suitably carried out in an open-air press of the type having a die(s) with a die wall defining a die cavity of suitable cross-sectional configuration. In such presses, the workpiece material or body is inserted in the die cavity and compacted or worked by opposing machine members, typically lower and upper punches. In the operation of such a two-punch press, the upper punch is initially raised out of the die cavity and the lower punch is initially in a low position so as to open the cavity to receive the material to be worked. The upper punch is then lowered to close the cavity, and the two punches are then mechanically or hydraulically actuated so as to press and compact the workpiece material between them. The punches closely fit the die wall so as to confine the material being worked but are slightly spaced from the die wall so as to reduce friction and wear. After the material is compacted, the upper punch is raised out of the cavity and the lower punch is raised so as to elevate the compacted workpiece above the top edge of the die or

so that the worked piece can be removed. This process is repeated on a more or less continuous basis.

In accordance with the broad context of the invention, a hot-pressed, fully-densified, permanent magnet body is produced in two pressing steps -- a cold-pressing step followed by a hot-pressing step. The present invention is practiced in the cold-pressing step.

Powder material of an above-described composition, in an amount based on the dimensions of the desired workpiece, is first compacted to a green compact body at ambient temperature and in air. This pressing can be called cold-pressing. The cold-pressed compact body suitably has a density of about five grams per cubic centimetre or higher, preferably about 5.3 to 5.5 grams per cubic centimetre. In this cold-pressing operation, a film of a solid die lubricant, such as Teflon<sup>™</sup> powder, is formed on the die wall of the press. No lubricant or binder is mixed with the rare earth element-containing powder.

The Teflon<sup>™</sup> or the like material is preferably applied in the form of a liquid suspension of powder in a non-flammable, highly volatile liquid vehicle. In this regard, it is preferable to use fully fluorinated aliphatic hydrocarbons of about two to eight carbon atoms per molecule. The fluid Teflon<sup>™</sup>-containing mixture is preferably applied to the die cavity wall through suitable small holes in the lower punch after the previously-formed compact body has been ejected from the die and the punch is being moved to its lowest position to receive the next charge of melt-spun powder. The upper punch is actuated to cold-press the powder into a porous green compact body. The dried lubricant film on the die wall facilitates the compaction and the removal of the compact body from the die without damage to the die or to the compact body.

After the green compact body has been formed, it is then ready to be hot-worked in another open-air press. Usually, a different press is employed because it is adapted to heat the die to facilitate the hot-pressing operation and requires heat-resistant tooling materials.

Some amount of liquid vehicle remains on the cold compact bodies. Presumably, this is due to the fact that no heat is applied to the press tooling and the pressing operation is carried out very rapidly. The solid lubricant film is not completely free of the liquid vehicle. Cold-pressed parts may not be hot-pressed immediately. Temporary storage of the compact bodies exposes them to both residual liquid vehicle and moisture-laden air. By using suitable volatile fluorinated hydrocarbon liquids as a vehicle for the formation of the solid lubricant film, it is possible to efficiently form rare earth-containing compact bodies that do not degrade on stor-

age.

Other objects and advantages of the invention will become apparent from a detailed description thereof, with reference to the accompanying drawings, in which:

Figures 1a to 1d are schematic views, partly in section, of a cold-forming, open-air press illustrating the sequence of cold compact forming-steps, including lubrication of the die cavity wall by spraying said wall with a liquid lubricant mixture supplied through the lower punch.

During the description of process of the invention, reference will be made to the drawings in which only a small portion of the press is depicted, namely that depicting the die and the upper and lower punches because it is in this region of the press that the special features of process of the invention are involved. A preferred embodiment of the invention is illustrated showing the making of a cold compact body intended to be hot-pressed into a sensor magnet in the shape of a circular right cylinder. However, it is to be understood that other magnet shapes can be produced by changing the die cross-section and punch shape. It is also to be understood that other press tooling constructions may be employed such as one punch anvil pressing, the pressing of ring shapes requiring cores, and the pressing of assemblies, i.e., magnets onto rotors or shunts, and the use of die shapes like shelf dies and step dies.

Figures 1a to 1d thus depict a small portion only of an open-to-the-air operable-at-ambient-conditions cold press 10. Cold press 10 has a die member 12 with a round cylindrical die cavity 14. Reciprocally operative in the die cavity 14 is a lower punch assembly 16. Also reciprocally operable in the die cavity is an upper punch 18. Upper punch 18 is slidably retained and guided by an upper punch carrier 20. Upper punch 18 has a round, flat punch face 22. As shown in Figures 1a to 1c, upper punch 18 has been raised to its uppermost position to facilitate removal of a compacted product from the die of the cold press and the addition of a new particulate starting material.

Lower punch 16 comprises a head 24, with a flat face 26, that is circular in cross section and adapted to closely fit the wall of die cavity 14, and a smaller diameter shank portion 28. Lower punch 16 also includes an enlarged base 30 that is below the die block 12. As shown in Figure 1a, the lower punch is elevated to its uppermost position with face 26 just flush with an upper surface 32 of die block 12. In this position, the lower punch has raised a just-formed cold compacted body of RE-TM-B particles 34. This cold compact body 34 has just been moved aside by a rake or other mechanical means (not shown) at the end of the compaction cycle of the press operation.

Typically such a cold compact body is a still slightly porous green compact body of RE-TM-B particles of the type described above. It has a density in excess of 5 grams per cubic centimetre and is very useful in accordance with the process for the hot-pressing and, if necessary, further hot-working of this compact body into a fully-densified magnet body with exceptionally good permanent magnet properties as disclosed in our aforesaid patent application.

Following the ejection of the cold compact body 34, lower punch 16 is then lowered to its lowest position (as shown in Figure 1b) in the operation of the press. It is during this lowering process that this lower punch carries out an important part of the practice of the present invention. Formed in lower punch 16 is a central axial duct 36 that extends from the base 30 of the punch 16 the length of the shank 28 of the punch and into the head 24. Axial duct 36 can be formed by drilling a hole through the base 30 up through the shank 28 into the head 24 and then closing off the outlet in the base with a plug member 38. Plug member 38 is preferably flush with the bottom of the base member 30 so that the mechanically-actuated press can operate on the bottom of the base to raise and lower the lower punch 16.

A transverse duct 40 is provided in the base member 30 that intersects axial duct 36. Duct 40 is threaded to receive fitting 42 and a supply tube 44 that is used for purposes that will soon be described. A small-diameter second transverse duct 46 with respect to axial duct 36 is drilled in the head 24 of the punch 16. The small duct 46 extends diametrically across the head 24 of the punch and has outlets in a machined annular ring 48 that is parallel to the face 26 of the punch but slightly below it at the upper end of axial duct 36. Thus, lower punch 16 contains a continuous internal passage leading from tube 44 into cross duct 40 through axial duct 36 to the small outlet duct 46 in the head 24 of the punch. The purpose of this passage is to supply a suitable lubricant to the wall surface of die cavity 14.

The use of a Teflon™ lubricant film is preferred. Teflon™ particles are applied by the use of a liquid carrier vehicle. The mixture is suitably about 90 percent by volume of the liquid vehicle and 10 percent by volume of Teflon™ particles. The liquid vehicle is a material that can suspend the Teflon™ particles if the liquid mixture is agitated and then carry them through the tube and ductwork of the lower punch. The liquid vehicle must also be a material that will readily vaporize from the wall of the die.

A suitable liquid vehicle for use in the present invention is a fully fluorinated derivative of an aliphatic hydrocarbon, preferably a hydrocarbon of 2

to 8 carbon atoms in the molecule. A perfluorinated hexane or octane is suitable. These molecules may be in the form of either molecular chains or the cyclo compounds. It is preferable to use perfluorinated hexane.

Thus, a mixture of about 90 percent by volume of liquid fluorocarbon and 10 percent by volume of Teflon<sup>™</sup> powder is mixed and prepared in a separate container not shown in the drawings. The mixture is agitated and then delivered from the container through tube 44 and ducts 40, 36 and 46 to the die cavity wall 14 of die 12. The container and delivery system (not shown) is adapted to supply the fluid under pressure as required.

Referring now to Figures 1a and 1b, the lubricant mixture is pressurized at the time that the lower punch 16 is at its uppermost point as depicted in Figure 1a. As the lower punch 16 is lowered in the die cavity 14 until it reaches its position as shown in Figure 1b, pressure is applied to the lubricant mixture and a coating film 50 of the lubricating mixture is applied to the cavity wall 14 of the die as depicted in Figure 1b. The liquid vehicle vaporizes very rapidly although there is a residual amount that remains in the film. Another important feature of the invention requiring the use of the perfluorinated compound is the fact that this material, if it remains on the surface of the cold compact body, does not adversely affect the permanent magnet properties of the body during any storage or subsequent hot-pressing thereof.

Thus, with the lower punch 16 in its down position and the upper punch 18 in its upper position and the lubricant film applied to the wall of the die cavity (Figure 1b), the cavity 14 is now ready to receive the powdered, rapidly-solidified iron-neodymium-boron type material. The lower punch 16 is raised to a fill position (Figure 1c) determined by the volume of powder to be added. The material is loaded into the lower die in loose particulate form. It is dropped into the die from a hopper (not shown), and it is measured by any suitable method into the die cavity. As seen in Figure 1c, the powdered material 52 is now in the die.

The upper punch 18 then comes down and the two punches 16, 18 and die wall co-operate with one another for the consolidation of the powder into the green compact body 34. Figure 1d illustrates the position of the upper and lower punches at the time that the particles have been consolidated into the green compact body 34.

As soon as the compaction has been completed, the upper punch 18 is raised out of the way to its upper position as depicted in Figure 1a, the lower punch 16 is raised to eject the compact body 34 from the die 12, the compact body 34 is removed, and the process is repeated. This cold-compaction process typically requires about one to

six seconds per cycle and is carried out at ambient conditions. The cold compact body will likely to have a trace of Teflon<sup>™</sup> powder on its outer surfaces. It will also have a trace of the fluorinated liquid vehicle. However, the composition of the liquid vehicle is such that it does not adversely affect the permanent magnet properties of the iron-neodymium type material, even though the compact body is stored in air before hot-pressing of the compact body takes place.

Whilst the practice of the invention has been described in terms of a few specific embodiments thereof, it will be appreciated that other forms of the invention could readily be adapted by those skilled in the art. As described, the lubricant film was formed by spray through one of the press punches. Obviously, the fluorinated liquid-powder mixture can be applied by other spray techniques, by wiping or similar coating procedure. Accordingly, the scope of the invention is to be considered limited only by the scope of the following claims.

#### Claims

1. A method of cold-pressing a rare earth element-containing alloy powder (52) of RE<sub>2</sub>TM<sub>14</sub>B precursor composition into a compacted body (34), preparatory to a hot-working operation on the compacted body (34), characterised in that the method utilises an open-to-the-air press (10) of the type comprising at least one die member (12) defining a material-receiving die cavity (14) with a die wall and opposing pressing members (16,18), at least one of which is adapted to move reciprocally in the die cavity (14), to compress material placed therein; said method comprises applying a solid lubricant film (50) to the cavity-defining wall which is at substantially ambient temperature, charging a predetermined quantity of a lubricant-and binder-free rare earth element-containing alloy powder (52) into the lubricated die cavity (14), and consolidating the powder (52) in the die cavity (14) by action of the pressing members (16,18) at ambient temperature to form a green compact body (34) of generally self-sustaining strength; and the solid lubricant is applied as a suspension of solid lubricant particles in a volatile, chlorine-free, fluorinated hydrocarbon liquid.
2. A method according to claim 1, in which the powder (52) is consolidated into a green compact body (34) of a density of about five grams per cubic centimetre or higher.
3. A method according to claim 1 or 2, in which

the die wall of the material-receiving die cavity (14) has a predetermined cross-sectional configuration, the opposing pressing members are upper and lower opposing punches (16,18) adapted to move reciprocally in the die cavity (14) to compress material placed therein, said predetermined quantity of a lubricant- and binder-free rare earth element-containing metal alloy powder (52) is charged into the lubricated die cavity (14) onto the lower punch (16), and the solid lubricant suspension is a suspension of solid lubricant particles in a liquid consisting essentially of a fully fluorinated aliphatic hydrocarbon of two to eight carbon atoms, which suspension is applied by spraying the suspension onto the die wall.

4. A method according to claim 1 or 2, in which the solid lubricant is applied to the die wall of the ambient temperature press (10) by spraying particles of polytetrafluoroethane or fluorinated ethylene-propylene copolymers dispersed in the fluorinated hydrocarbon liquid through a duct (36,46) in the lower punch (16).
5. A method according to claim 3, in which the solid lubricant is applied to the die wall of the ambient temperature press (10) by spraying particles of polytetrafluoroethane or fluorinated ethylene-propylene copolymers dispersed in the fluorinated hydrocarbon liquid through a duct (36,46) in the lower punch (16).

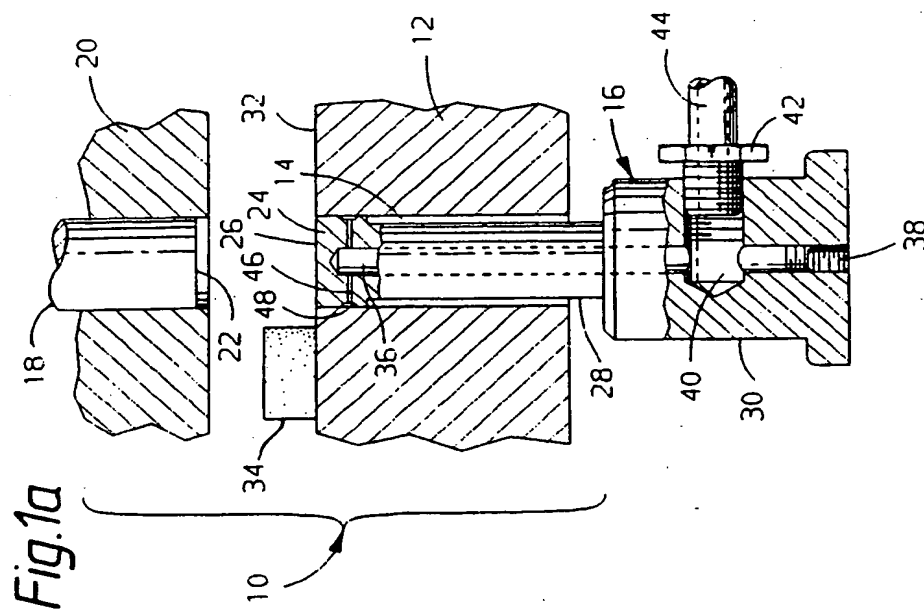
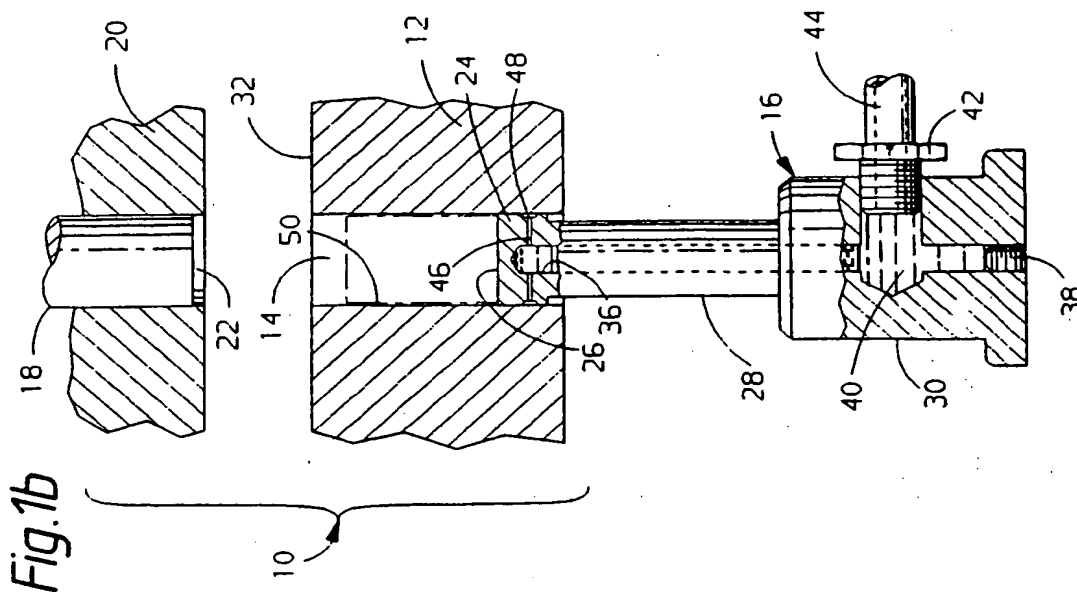
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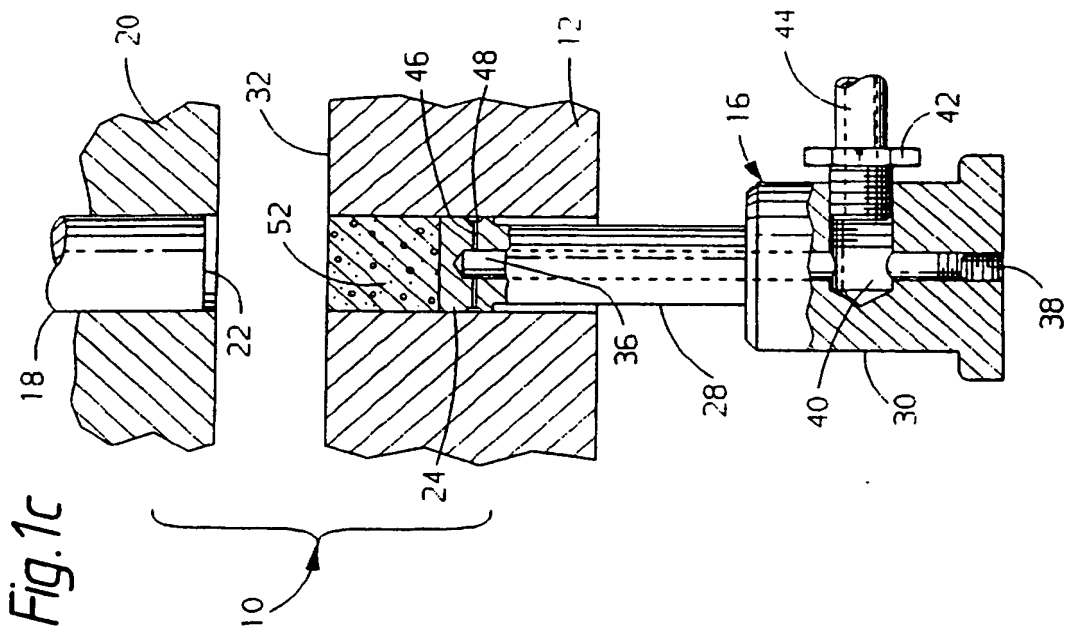
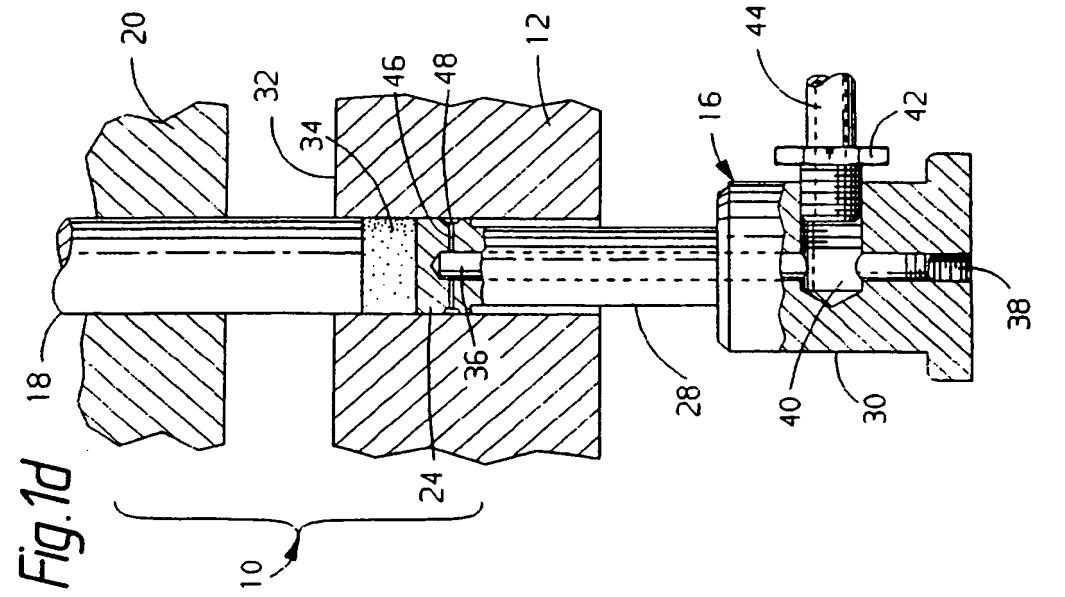
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## EUROPEAN SEARCH REPORT

Application Number

EP 92 20 1202

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
A	GB-A-2 235 700 (GEC ALSTHOM) * page 2, paragraph 3 - page 3, paragraph 1; claims 1,7 *	1	H01F1/053 H01F41/02
A	EP-A-0 302 598 (GENERAL MOTORS CORP.)		
A	PATENT ABSTRACTS OF JAPAN vol. 13, no. 246 (E-769)(3594) 8 June 1989 & JP-1 048 407 ( SEIKO EPSON CORP. ) * abstract *		
			H01F
The present search report has been drawn up for all claims			
Place of search <b>THE HAGUE</b>		Date of completion of the search <b>11 AUGUST 1992</b>	Examiner <b>DECANNIERE L.</b>
<div style="display: flex; justify-content: space-between;"> <div> <p><b>CATEGORY OF CITED DOCUMENTS</b></p> <p>X : particularly relevant if taken alone</p> <p>Y : particularly relevant if combined with another document of the same category</p> <p>A : technological background</p> <p>O : non-written disclosure</p> <p>P : intermediate document</p> </div> <div> <p>T : theory or principle underlying the invention</p> <p>E : earlier patent document, but published on, or after the filing date</p> <p>D : document cited in the application</p> <p>L : document cited for other reasons</p> <p>.....</p> <p>&amp; : member of the same patent family, corresponding document</p> </div> </div>			

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